



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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Applicant : Ouyang, Michael  
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Title : OLED Structures with Strain Relief,  
Antireflection and Barrier Layers  
Art Unit : 2879  
Examiner : Hines, Anne M.  
Docket No. : SP03-075

Commissioner for Patents  
P.O. Box 1450  
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CERTIFICATE OF MAILING (37 CFR 1.8a)

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January 30, 2006

William S. Francis J. McRiddell

DECLARATION UNDER 37 C.F.R. § 1.131

1. I, Michael X. Ouyang, am an inventor of the claimed subject matter of US Patent Application No. 10/698,723, Corning attorney docket no. SP03-075. I declare that I have first-hand knowledge of the facts set forth in this Declaration.
2. The invention claimed in the referenced patent application was conceived and reduced to practice in the USA.
3. I understand that willful false statements and the like contained herein are punishable by fine, imprisonment, or both, as provided by 18 U.S.C. § 1001, and may jeopardize the validity of the application or any patent issuing thereon.
4. The invention claimed in the referenced patent application was conceived on a date prior to March 29, 2002 and diligently reduced to practice as a laboratory model beginning at least as early as March 29, 2002. Conception is evidenced by the attached pages from my "Laboratory Notebook" and supporting description prepared after the actual reduction to practice of the invention claimed in the reference U.S. Patent Application. The attached document is a business document generated in the course of technological research and development as part of documenting inventions discovered in the regular course of business.

I declare that all statements made herein are based upon my own knowledge and to the best of my knowledge are true and all statements made on information and belief are believed to be true.

Michael X. Ouyang

1/20/2006  
Date

OLED Substrate

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## Flexible OLED Substrates with Sun-light legible and anti-scratching

### Background

Sunlight legible is important for displays, especially handheld displays, such as cellular phones, digital and video cameras, automobile displays, where the display could be exposed to the very intense sun-light. The bright light could "wash off" the information displayed. To improve the contrast ratio of the display, dark background was proposed. Dark metal films for rear electrode were reported by Takeda<sup>[1]</sup>. These metal includes Mo, Zr, Ti, Y, Ta, Ni, Al with thickness of 1-300Å. Dobrowolski et al described "an optical interference, electroluminescent device having low reflectance using multilayer metal/dielectric combination as the rear electrodes<sup>[2]</sup>". In these inventions, the low reflective components are the patterned electrodes.

Flexible substrate is very attractive for the next generation flat panel displays, such as organic light emitting diodes (OLED). The possible candidate for flexible substrates are polynorbornene (Tg:320 °C), polyimide (Tg:270-388 °C, DuPont's Kapton), polyethersulphone (Tg: 184-230 °C), polyetherimide (Tg: 204-299 °C), polyarylate (Tg:148-245 °C), polycarbonate (Tg: 150 °C), Transphan (Tg: 171 °C). However, OLED performance is well known to be sensitive to humidity and oxygen. The above polymer substrates must be modified for OLED applications. The state of art packaging technique to enhance the barrier property of polymer substrates includes a) inorganic coatings on polymer substrate by PECVD<sup>[3]</sup>; b) organic/inorganic laminar structure<sup>[4]</sup>; c) Multifunction (filter, polarizing plate, phase difference plate, viewing angle control plate, LC alignment ...) laminated \_hard pressing<sup>[5]</sup>; d) Atomic mixing of F doped multilayer deposition of polymer composites by flash-evap and high energy radiation source (electron, UV, IR, microwave, ultrasonic, or gama radiation)<sup>[6]</sup>.

Inorganic coatings has low yield strength, it could crack during the substrate bending. On the other hand, the hermetical properties of the coated polymer substrates depend on the coating thickness. But a thick dielectric coating will induce substrate curving due to the residual stress in the coatings. In this invention, we propose a coating structure for the rear and front flexible substrates by using a combination of metal and hard inorganic coatings. The New OLED substrate has a high yield strength, a low or high reflectivity depending on the application and an anti-scratching at the front surface.

A typical structure of OLED is described in figure 1 (<http://www.uniax.com/>), where the top and the bottom substrates can be glass or polymers. The front substrate (bottom) must be transparent and the rear substrate does not need to be transparent. The cathode lines are low work function materials for electron injection, such as Ca, Li, Mg or their alloy Mg/Ag, Al/Li or multilayer LiF/Al, Li<sub>2</sub>O/Al, CsF/Al structures. The anode material must be transparent for light pass through. ITO with high work function modified surface are required. In our invention, The surface of the front substrate will be coated with stress compensated multi-dielectric layers for antireflection, anti-scratch and humidity

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barrier coatings and the back side the the rear substrate will be coated with metal/dielectric combination as the dark or mirror humidity barrier coatings. Detail is shown in figure 2.

Nanocomposite is a good hydrophobic and anti-scratching layer.

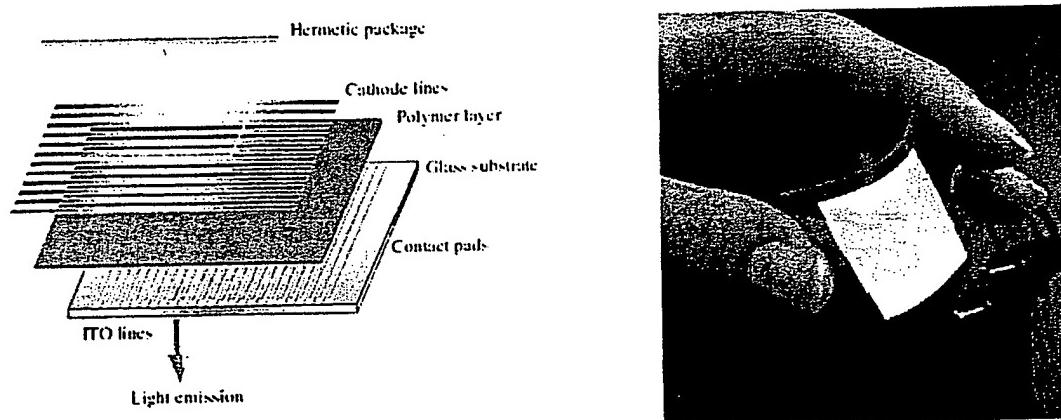


Figure 1. OLED device structure by Uniax.

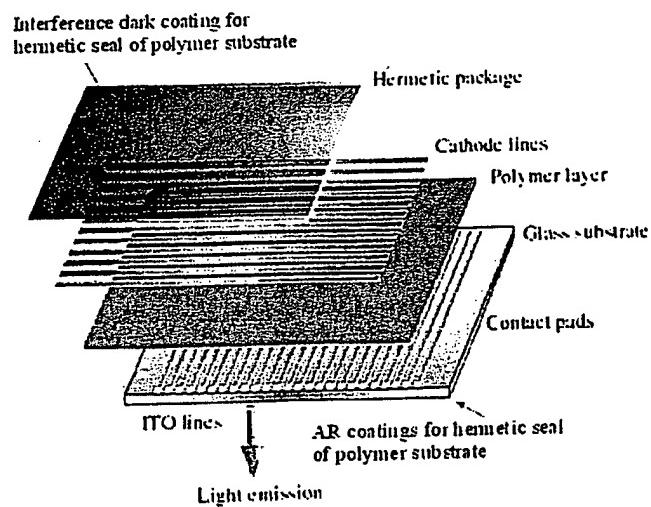


Figure 2. Our invention of modified flexible substrates with good water and oxygen barrier properties and a AR nad high contrast properties.

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OLED substrate

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E \_\_\_\_\_

Technical DescriptionDark coatings for rear substrate

The structure of humidity and oxygen barrier layer is shown in figure 3.

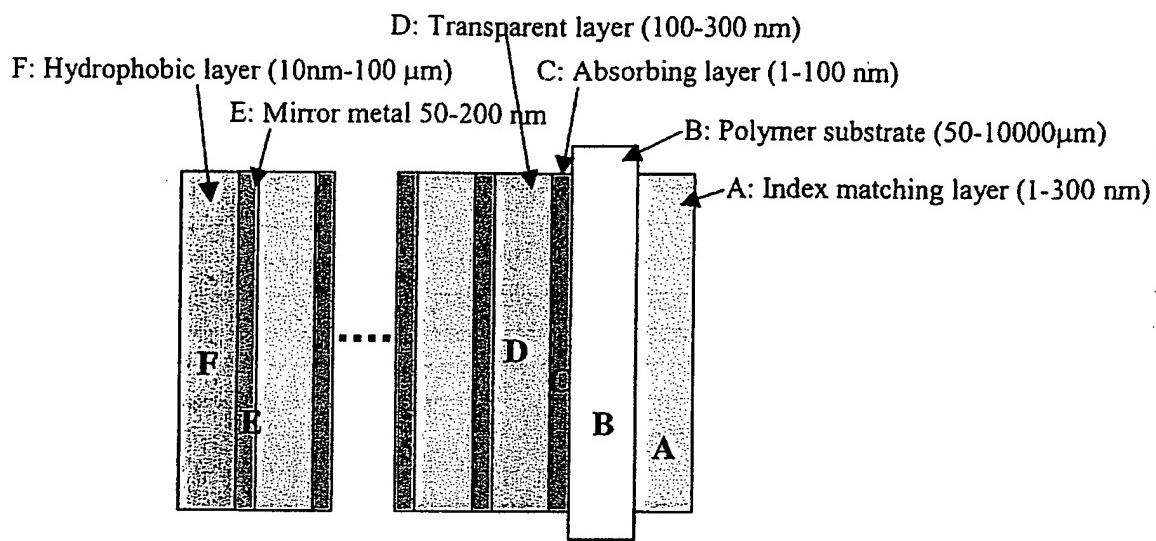


Figure 3. Coating structure on a rear polymer substrate of a OLED

The dark coatings can be constructed by metal/dielectric stacks with interference structure. The theory is based on the well known interference phenomena that the light intensity will be canceled if the reflected light from difference thin film interfaces/surface has a added equal intensity but opposite in phase (or 180 degree out of phase).

The absorbing layer C can be a thin dark metal coating (Mo, Zr, Ti, Y, Ta, Ni, W, Inconel, etc,) or thin absorbing dielectric(diemond-like Carbon, SiO<sub>x</sub>, oxygen deficient In<sub>2</sub>O<sub>3</sub>, ITO, SnO<sub>2</sub> etc) or a semiconductor coating (Si, Se, Ge, GaAs, GaN, Se, GaSe, GaTe, CdTe, TiC, TiN, ZnS, ZnO, CdSe, InP, BN, etc.)

The transparent layer can be dielectric layer with quarter wavelength thickness (Al<sub>2</sub>O<sub>3</sub>, AlON, BaF<sub>2</sub>, BaTiO<sub>3</sub>, BeO, MgO, GdO<sub>3</sub>, Nb<sub>2</sub>O<sub>5</sub>, ThO<sub>2</sub>, CeO<sub>2</sub>, HfO<sub>2</sub>, Sc<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub>, Si<sub>3</sub>N<sub>4</sub>, TiO<sub>2</sub>, Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>, Y<sub>2</sub>O<sub>3</sub>, ZeSiO<sub>4</sub>, Ta<sub>2</sub>O<sub>5</sub>, HfN, ZrN, AlN, SiC, Bi<sub>12</sub>SiO<sub>20</sub>, etc)

Coating C and D are the basic repeating structure to enhance the oxygen and humidity barrier effect. The thickness of layer E may be different from stack to stack. Coating A and/or F may or may not necessary depending on the structure and the materials performance.

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The mirror metal can be Al, Au, Ag, Pt, or dielectric mirrors.

The above materials can be deposited <100 °C by e-beam or by sputtering or by web coating techniques.

Figure 4- figure 5 is the simulation of reflectivity of light from multilayers through polymer (Topas® (n=1.53)) or glass (n=1.52-1.53) substrate.

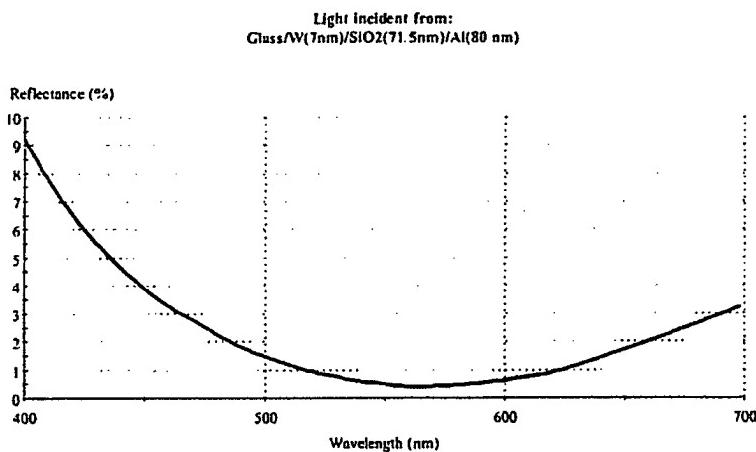


Figure 4 Reflection of three layer structure where light is reflected from Topas® or glass/W(7nm)/SiO<sub>2</sub>(71.5nm)/Al(80 nm)

Light Incident from:  
Glass/W(6.1nm)/SiO<sub>2</sub>(78.5nm)/W(15.3nm)/SiO<sub>2</sub>(78.5)/Al(71 nm)

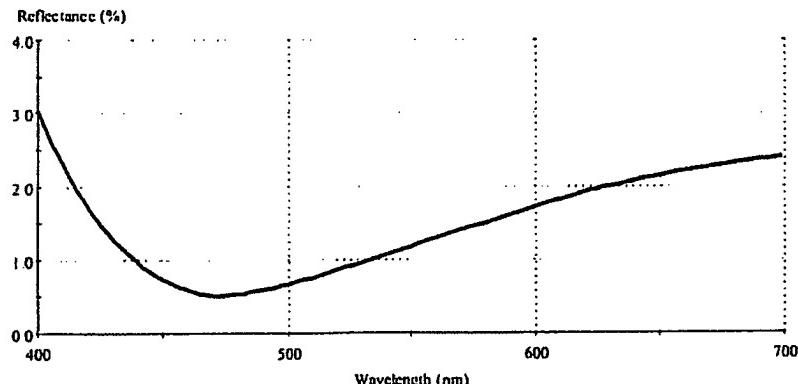


Figure 5 Reflection of three layer structure where light is reflected from Topas® or glass/W(6.1nm)/SiO<sub>2</sub>(78.5nm)/W(15.3nm)/SiO<sub>2</sub>(78.5)/Al(71 nm).

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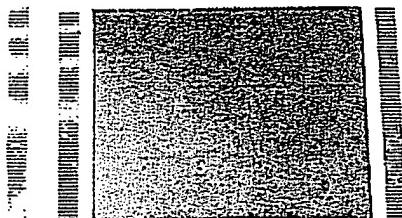
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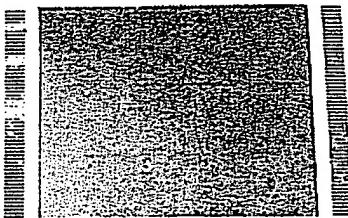
Experimentally, we demonstrated the antiglare effect in figure 6 and figure 7. Figure 7 shows that a dark metal electrodes with combination of dark structure, the display has no anti-glare and is sunlight legible.



(a)

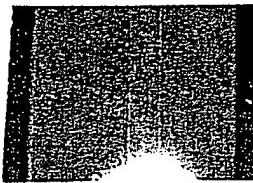


(b) Al(150nm) electrodes on glass

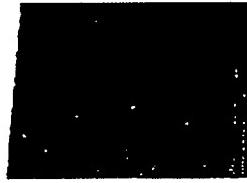


(c) W(20nm)/Al(100nm) electrodes

Figure 6. (a) Dark multiplayer on glass, (b)Al electrode on glass, and (c) black electrode on glass.



Structure (b) On (a)



Structure (C) on (a)

Figure 7. structure (b) on (a) and structure (c) on (a) of figure 5

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PURPOSE

We claim using stress compensation technique to minimize the stress of film stack induced substrate curving. By control the process, such as sputtering pressure, deposition rate and the type of material.<sup>[7, 8, 9]</sup> We can achieve zero total stress. Another way to prevent the polymer from warping is to coat inorganic materials on both sides of the polymer substrate to balance the stress.

We claim a hydrophobic materials as the outer layer to cover the whole optical and oxygen barrier. The hydrophobic layer can be polymer layer, such as PTFE, nylon, glass fibers, and polyethersulfone, as well as nano-composite clays such as polymer-layered silicate nanocomposites.<sup>[10]</sup> These serious of nano materials can greatly enhance the moisture barrier property as well as the hardness for surface anti-scratching.

## AR coating on front substrate

One to four inorganic layers can be used as the oxygen and humidity barrier layer. Figure 8 shows a three layer antireflection structure. The hydrophobic layer should have a refractive index less than 1.45. transparent inorganic layers with two or three different refractive indexes are required for multilayer AR design.

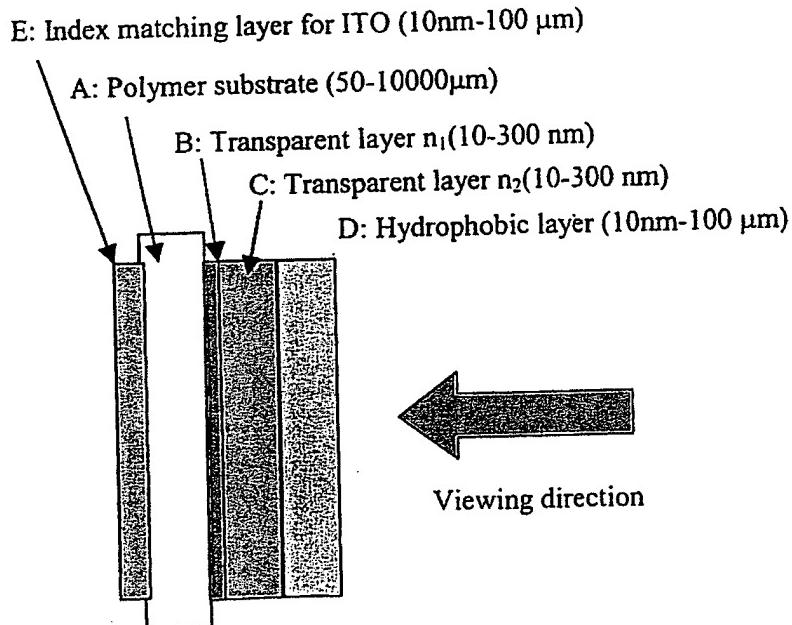


Figure 8. A typical three layer AR design where the outer layer could be a hydrophobic materials with low refractive index.

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The refractive index of one layer AR coating should be close to  $n_{\text{substrate}}^{1/2}$ . ITO has a refractive index of about 2.0 at 550 nm. The index matching layer E should have a  $n \approx 1.81$ .  $\text{Si}_3\text{N}_4$ ,  $\text{SiON}$ ,  $\text{BiO}_2$ , etc are the best choice.

Multilayer AR coating give a broad AR band as well as a better barrier effect. The choice of each layer depending on the refractive index and thickness. For a three layer AR coating, Angus Macleod described that the condition for the vectors to be equal length but opposite directions should be<sup>[11]</sup>:

$$y_1/y_0 = y_2/y_1 = y_3/y_2 = y_{\text{sub}}/y_3$$

Where  $y_i$  ( $i=0,1,2,3,$ ) is the optical admittance of the  $i$ th layer. 0 is referred to the air. Therefore, if  $n_{\text{substrate}} = 1.52$ , the four layer AR coating can be air/MgF( $n=1.38$ , 92.7 nm)/ZrO<sub>2</sub>( $n=2.06$ , 131.7 nm)/MgF(30.3 nm)/ ZrO<sub>2</sub>(16.5 nm).

Other high/low (refractive index) combination using the transparent dielectric films as described in previous section is also possible.

nano-composite clays can also be used for the first layer for water barrier as scratch resistant coating.

### References

- <sup>1</sup> M. Takeda et al: US Patent: 4,287,449, 1981
- <sup>2</sup> EP 0372763A2
- <sup>3</sup> WO 01/82389 A1
- <sup>4</sup> EP 0977469A2, WO 00/36665
- <sup>5</sup> US6,217,995 B1, US4,647,508
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- <sup>7</sup> H. Kattelus , et al: "Stress control of sputter-deposited Mo-N films for micromechanical applications", Microelectronic Engineering 60 (2002) 97–105.
- <sup>8</sup> S. Stadler, et al: "Stress control in thin sputtered films towards potential application in micromachined structures", Materials Letters 35 1998 18–21
- <sup>9</sup> Marcia C.K. Tinone, et al: "Multilayer sputter deposition stress control", J. Electron Spectr. & Related Phenomena 80 (1996)461-464.
- <sup>10</sup> Michael Alexander, Phillippe Dubois, : Polymer-layered silicate nanocomposites: preparation, properties and uses of a new class of materials." Materials Sci. & Engin., 28 (2000)1-63.
- <sup>11</sup> H. Angus Macleod, "Thin-Film Optical Filters" 3<sup>rd</sup> Edition, 2001, P102.

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